

FWP and possible subtask under FWP:

Integrated Modeling of Novel Materials

FWP Number:

SCPE420

Program Scope:

We have refocused the theory effort to study short time and length scales with respect to the static and dynamic properties of materials. These two major directions are closely connected and represent complimentary and necessary aspects of a theoretical effort on nanotechnology. This redirection, which is a natural extension of our previous work, closely aligns us with the new Los Alamos and Sandia Center for Integrated Nanotechnologies (CINT), where state of the art probes of matter such as scanning tunneling microscopes (STM), time-resolved scanning tunneling microscopes, near field scanning optical microscopes (NSOM), and other local and fast probes will be developed and used. This work involves an attempt to understand the dynamical properties of local structures that are measured by these novel probes, and how to exploit their unique functionalities. These local structures arise either because they are deliberately engineered, such as quantum dots, or are inherent due to competing interactions. Thus, this work will include the study of local electronic properties of strongly correlated materials and artificially created nanostructures, such as quantum dots, metal nanoparticles on DNA, impurity and vibronic states, Inelastic Electron Tunneling Spectroscopy (IETS), fast optical response of these materials, and electron-phonon coupled systems such as colossal magnetoresistance (CMR) manganites. We will further develop basic theoretical methods that will make possible detailed analysis and predictions about specific experimental properties.

Major Program Achievements (over duration of support):

- Developed a microscopic theory of the STM detection of the bosonic collective modes in high-T_c materials.
- Developed a description of the dynamics of a single spin coupled to a Josephson current in the presence of the ac Josephson effect.
- Developed a microscopic description of the coupling between atomistic lattice distortions and the electronic properties of unconventional superconductors.
- Calculated the essentially exact quantum dynamics of the formation of a polaron quasiparticle.
- Measured and explained coupled charge-spin dynamics in magnetoresistive pyrochlore $\text{Ti}_2\text{Mn}_2\text{O}_7$.

Program impact:

Applied advanced modeling and simulation tools to interpret and guide modern experimental probes of multiple spatial and temporal scales, including high magnetic fields, ultrafast time-resolved optical and vibrational spectroscopies, and scanning-tunneling microscopies.

Interactions:

Internal: J.D. Thompson, E. Bauer, T. Park (MST-10) – heavy fermion Ce115 compounds, T Taylor, R. Averitt, R. Prasanumar(MST-CINT), S. Crooker (NHMFL) – optics, S. Tretiak (T-12) – molecular electronics, Q. Jia(MST-STC) – multiferroics.

Several external collaborators: J.C. Davis (Cornell), E. Abrahams(Rutgers), T. Egami (Oak Ridge), P. C. Hammel (Ohio State), P. Littlewood (Cambridge), Allan H. MacDonald (UT Austin), Q. Si (Rice), L. Ku(Texas), J. Bonca and J. Demsar (Ljubljana).

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

A.V. Balatsky- LANL Lab Fellow, Oct 2005.

Personnel Commitments for FY2005 to Nearest +/- 10%:

A.V. Balatsky – 30%

S.Trugman – 30%

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 200k

FY04 BA \$ 170k

FY05 BA \$ 150k

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0201020

FWP and possible subtask under FWP:

Nanophosphors: Fundamental Science of Insulators in the Nanoscale Regime

FWP Number: SCPE972

Program Scope:

The goal of this program is to assess the effects of reduced dimensionality on inorganic, insulating phosphors. Size reduction of high-quality bulk phosphors to nanoscale dimensions ("nanophosphors") is expected to alter structural, optical and magnetic properties due to the resulting large surface-to-volume ratio and concomitant overlap of dopant-ion/lattice-ion wavefunctions. Scope of the work consists of three major components: (1) fabrication of nanophosphors based on well-characterized bulk phosphors; (2) nanophosphor characterization by structural, optical, and magnetic resonance techniques; and (3) interpretation of results vis-à-vis bulk properties. The ultimate goal is to understand the role of reduced dimensionality so that nanophosphors may be tailored to meet technical demands for new materials in radiation detection and lighting applications.

Major Program Achievements (over duration of support):

Nanophosphor $\text{Y}_2\text{SiO}_5\text{:Ce}$ (*n*-YSO) was prepared by the hydrothermal technique and examined by optical and structural methods; significant deviations from bulk properties were found. *n*-YSO crystallized in the $P2_1/c$ structure (7 & 9 oxygen coordination) rather than the expected $B2/b$ structure (6 & 7 oxygen coordination) observed in bulk material. This is surprising because calculations based on density functional theory suggest that the latter structure is more energetically favorable. Relative to bulk powder, *n*-YSO exhibited red shifts of the photoluminescence excitation and emission spectra, reduced self-absorption (larger Stokes shift), enhanced light output (factor of 3), and medium-dependent radiative lifetime. Photoluminescence data are consistent with reduced symmetry of the $P2_1/c$ structure and are not necessarily related to reduced dimensionality of the nanophosphor. In contrast, medium-dependent lifetime and enhanced light output are directly attributed to nanoscale behavior. Perturbation of the local electric field of the Ce ion is responsible for the medium-dependent lifetime. Interestingly, this latter effect provides a new and novel technique for extracting electronic transition oscillator strengths. This work has been recently submitted for publication in Applied Physics Letters (D. W. Cooke, *et al.*, "Luminescent properties and reduced dimensional behavior of hydrothermally prepared $\text{Y}_2\text{SiO}_5\text{:Ce}$ nanophosphors").

Program impact:

Initial research has confirmed our expectations of new and novel behavior in nanophosphors and has laid the foundation for a research program focused on exploring these intriguing reduced-dimension effects. Already the present data suggest that nanophosphors offer the possibility for tailored spectral emission and excitation, variable radiative lifetime, and enhanced light output—technologically important properties that cannot easily be altered in bulk phosphors. Implications for improved radiation detectors based on these results have been realized via a patent application, an invention disclosure, a non-disclosure agreement with Reuter-Stokes Radiation Measurement Solutions (GE Energy) and inquiries from several companies regarding this technology.

Interactions:

J.-Y. Kim and K. S. Hong (Seoul National University); L. Clark and J. Williams (Reuter-Stokes); Steve Duclos (GE Global Research); J. Phillips (Univ. New Mexico); M. Jahan (Univ. Memphis). Internal LANL Interactions: K. Ott, Y. Wang, P. Arendt, M. McClesky, E. McKigney.

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

R. E. Muenchausen, D. W. Cooke, J.-K. Lee, M. Nastasi and Q. Jia, "Method for Preparation of Rare-Earth Oxyorthosilicate Phosphor Films." S-104,811 (patent pending).
D. W. Cooke, E. A. McKigney, R. E. Muenchausen and B. L. Bennett, "Nanocomposite Scintillator and Detector." S-104,966 (invention disclosure filed Aug. 1, 2005).
Conversion of L. G. Jacobsohn from postdoctoral status to technical staff member now working on the BES-funded program.

Personnel Commitments for FY2005 to Nearest +/- 10%: (Program began in March '05)

W. Cooke (35%); R. Muenchausen (20%); B. Bennett (30%); J. Smith (10%); M. Nastasi (5%); Q. Jia (5%); K. Sickafus (5%); J.-K. Lee (25%); L. Jacobsohn (5%); B. Uberuaga (15%); J. Valdez (10%).

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 0

FY04 BA \$ 0

FY05 BA \$ 450k

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0201020

FWP and possible subtask under FWP:

Deformation Physics of Ultrafine Scale Materials

FWP Number: SCPE486

Program Scope: This program investigates the unusual deformation physics being discovered in ultrafine scale materials that lead to strength levels near the theoretical limit of strength for perfect crystals. The program involves a synergistic combination of atomistic simulations and experimental methods. Current focus is primarily on nanolayered metallic composites. The integrated approach consists of synthesis by vapor deposition; structure-property correlations by means of transmission electron microscopy, x-ray diffraction, microtensile and nanoindentation testing; and atomistic simulations of deformation behavior.

Major Program Achievements (program started in FY00, with full funding in FY01):

Nanoscale design of metals with strengths approaching the theoretical limit: An increase of up to two orders of magnitude in strength over bulk materials is observed when the structural scale of the nanolayered composites is reduced to 1-2 nanometers.

New Strengthening Mechanisms in Nanolayered Materials: Atomistic modeling reveals a new strengthening mechanism in incoherent interfaces; an unusually high resistance to slip transmission originates from dislocation-induced shear of 'weak' interfaces and concomitant dislocation core spreading within the interfaces. We find that coherent multilayers derive their high strengths primarily from coherency stresses.

Unusual thermo-mechanical stability develops - composites deform homogeneously to large plastic strains while preserving the crystallography. The structure has been found stable at unusually high temperatures.

Nanoscale twinning was discovered in certain single component systems (e.g., austenitic stainless steels) resulting in increase in strength levels similar to multi-component layered systems.

Program impact: This program has discovered new regimes of plasticity in nanoscale composite materials that have led to new scientific models that help to understand the origins of mechanical behavior of nanoscale materials as their strength approaches theoretical limits. The work from this program is largely the basis for the nanomechanics thrust area in the upcoming Center for Integrated Nanotechnologies in New Mexico. The scientific knowledge from this program promises to impact a broad range of engineering applications including load-bearing structural components, components subject to wear, MEMS, and high strength electrical conductors.

Interactions (FY'05):

Hussein M. Zbib (Washington State University), Peter M. Anderson (Ohio State University), Amiya K. Mukherjee (UC, Davis), Ian M. Robertson (University of Illinois), Tamas Ungar (Eotvos University Budapest, Hungary).

Recognitions, Honors and Awards (since FY'00):

R.G. Hoagland, LANL, Matthias scholar at LANL, FY'00.

P.M. Anderson, Ohio State University, Matthias scholar at LANL, FY'02.

J.D. Embury, 2005 ASM Edward DeMille Campbell Memorial Lecture.

F. Spaepen, 1999 Humboldt Research Award for Senior U.S. Scientists.

F. Spaepen, 2002 Robert Franklin Mehl Award, The Minerals, Metals & Materials Society (TMS).

60 refereed publications, including 3 book chapters; 25 invited talks at national/ international symposia.

8 symposia organized (MRS Fall 2000; United Engineering Foundation, Italy, 2001; TMS Annual 2003; MRS Spring 2003; TMS Annual 2005; MRS Spring 2004; MRS Spring 2005; MRS Spring 2006). Edited 2 journal special issues: Scripta Materialia issue titled *Deformation and Stability of Nanoscale Metallic Multilayers* (March 2004), and MRS Bulletin issue titled *Mechanical Properties of Nanostructured Materials*, 1999.

Personnel Commitments for FY2005 to Nearest +/- 10%:

R.G. Hoagland (LANL, 35%), A. Misra (LANL, 35%), F. Spaepen, (Harvard University, 5%), J.D. Embury (LANL-Affiliate, 10%), J. P. Hirth (LANL-Affiliate, 10%), S.G. Srinivasan (LANL TSM, 20%), J.G. Swadener (LANL TSM, 10%), X. Zhang (LANL post-doc, 50%), T. Hoechbauer (LANL post-doc, 50%), A. Donohue (100%, GRA, Harvard University), F. E. Akasheh (50%, GRA, Washington State University), K. Hattar (summer GRA).

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$759K

FY04 BA \$759K

FY05 BA \$729K

Laboratory Name: Los Alamos National Lab
B&R Code: KC0202010

FWP and/or subtask Title under FWP:
Photoelectron Spectroscopy of Transuranics

FWP Number: SCPE355

Program Scope:

Electronic structure investigations of transuranic and actinide materials using photoelectron spectroscopy including angle-resolved (ARPES) and resonance photoemission. Development of a plasma-based tunable photon source for transuranic photoemission. Synchrotron-based research for uranium and lighter materials using ARPES.

Major Program Achievements (over duration of support):

Photoemission results for 10 transuranic compounds, Np metal and 2 phases of Pu metal. Detailed photoemission and model calculations for delta phase Pu metal indicating the 5f electrons for Pu are founding both in a localized and itinerant configuration (J. Electron Spectrosc. Relat. Phenom. 135, 163 (2004)). Clear demonstration of band magnetism in UTe by angle-resolved photoemission (Phys. Rev. Lett. **93**, 267205 (2004)). A comprehensive overview of actinide tellurides and antimonides (Phys. Rev. B 70, 205103 (2004)). A detailed study of oxide and hydride formation on delta phase Pu metal surfaces (Surface Science 571, 74 (2004)). First photoemission measurements on the Pu superconductor PuCoGa₅ with mixed-level-model calculations in excellent agreement with the photoemission (Phys. Rev. Lett. 91, 176401 (2003)). Angle-resolved photoemission for USb₂ showing the narrowest 5f bands ever reported (Phys. Rev. B 69, 45102 (2004)). Overview of correlated electron spectroscopy in terms of narrow f-electron bands (J. Electron Spectrosc. Relat. Phenom. 117, 323 (2001)). Determination of the first order phase transition and ramifications for the electronic structure in YbInCu₄ (Phys. Rev. B 63, 7101 (2001); Phys. Rev. B 62, 16492 (2000)). Direct comparison between PES and calculations for α and δ -Pu (PRB 62, 1773 (2000)). First measurement for Np compound hybridization and 5f bands (Los Alamos Science 26, 120 (2000)). Demonstration of band behavior for actinide f-electrons (Gschneidner Handbook, Chap. 172 (1999)). LPLS design and laser ablation for cleaning transuranics (J. Alloy & Comp. 286, 14 (1999)). First resonance photoemission data on Pu metal (Surf. and Interface Analysis 26, 121 (1998)). Design, construction, testing of the laser plasma light source (1992-1996).

Program impact:

One of two programs world-wide to provide photoemission data on transuranic elements and compounds. Experimental results directly impact model calculations and the current thinking for transuranic materials. The only facility in the world with tunable photons for vacuum-ultra-violet or soft x-ray photoemission of transuranics.

Interactions:

External – Ames Lab – Cliff Olson
Synchrotron Radiation Center – Hartmut Hochst
Institute for Transuranium Elements, Karlsruhe, Germany – Gerry Lander
Uppsala University, Uppsala, Sweden – Olle Eriksson, Peter Oppeneer
Internal – Nuclear Materials Technology division – Luis Morales, Theory division – John Wills

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

Joyce, Arko and Durakiewicz have had 32 invited talks 42 publications (1997 to present) including 2 book chapter. Joyce, Arko and Graham received the Los Alamos Distinguished Performance award for this project 1997. J.J. Joyce elected to users advisory committee of Synchrotron Radiation Center and currently chairs the committee (1999-present, chair 2003-present); chair 36th SRC meeting Oct. 2003; co-chair MRS symposium on Actinides Dec. 2003; local arrangements committee 61st Physical Electronic Conference 2001. A.J. Arko elected Fellow of Los Alamos National Lab 1999.

Personnel Commitments for FY2005 to Nearest +/- 10%:

John Joyce (PI) 20%; Tomasz Durakiewicz (TSM) 20%; Postdoc 50%, Kevin Graham (TEC 7) 30%

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$310,000

FY04 BA \$300,000

FY05 BA \$300,000

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0202030

FWP and/or subtask Title under FWP:
NHMFL 100T Multi-Shot Magnet

FWP Number: SCPE438

Program Scope: The objective of this activity is to design, construct, and operate a 15mm bore, 100T Multi-Shot (MS) magnet to be installed at the NHMFL Los Alamos Pulsed Field Facility. Construction is underway and we have accomplished major milestones during 2005.

Major Program Achievements (over duration of support):
Successfully tested and operated the 4MJ insert capacitor bank

Successfully tested 75T insert prototype

All outer coil set (7 in total) completed

Preparation for ABB Power Supply modifications underway

Program Impact:

The 100T-MS magnet is a multidisciplinary project involving many different aspects of materials design, materials research, high-power systems, and unique instrumentation design. This project benefits from the high-power design expertise of many groups across LANL, and the expertise in instrumentation innovation at the NHMFL-LANL. Under the NHMFL – LANL management, a team of engineers at LANL – ESA-WDS is responsible for designing and constructing the outsert coils (7 in total). The short-pulse insert coil is under development in collaboration with engineers of the NHMFL site at Florida State University, while LANL science teams are preparing new measurement processes required for this unique magnet environment.

Interactions:

NHMFL, Florida State University
LANL, ESA-WDS
Everson-Tesla; Large Coils construction
ABB; Power Supply

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

4 Invited talks and one Award related to the 100T Project
Lacerda; Tsukuba-Japan, March 2005
Lacerda; ISSP-Japan, March 2005
Lacerda; Dresden-Germany, November 2005
Lacerda; Toulouse-France, November 2005
LANL Small Team Award: 4Mj Capacitor Bank and 75T magnet testing

Personnel Commitments for FY2005 to Nearest +/-10%:

J. Sims (TSM-LANL-ESA; Eng.) 15%	A. Paris (TEC-LANL-NHMFL) 50%
C. Ammermann (TSM-LANL-ESA; Eng.) 25%	K. Kihara (TEC-LANL-ESA) 50%
D. Rickel (TSM-LANL-NHMFL) 25%	M. Gordon (TEC-LANL-NHMFL) 50%
G. Ellis (TSM-LANL-ESA; Eng.) 25%	J. Schillig (Affiliate/TSM-LANL-NHMFL; Eng.) 50%

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 665k

FY04 BA \$ 665k

FY05 BA \$ 630k

FWP and/or subtask Title under FWP:

Ion-Enhanced Synthesis of Materials

FWP Number:

SCPE407

Program Scope: To develop a fundamental understanding of ion and plasma processed materials and to determine how ion and plasma processing parameters influence the material structure and functional properties. Experiments, theory and modeling are used to understand the underlying physics and fundamental processes responsible for the enhanced properties derived by materials synthesis by these methods.

Major Program Achievements (over duration of support): We have successfully developed the Plasma Immersion Ion Process (PIIP) to synthesize diamond-like-carbon (DLC) field emitters. Modeling showed that electron emission is dependent sp^3 contents and the aspect ratio of the emitting area. PIIP was then used to alter the surface topology and ratio of sp^2 to sp^3 carbon bonding at the surface and thereby control the electron emission behavior.

We have shown that the intrinsic stress in thin films formed by ion assisted deposition techniques are strongly influenced by the presence of grain boundaries, vacancies, interstitials and dislocations. An atomic interaction model has been developed to explain the source of intrinsic stress in thin films.

We have shown for the first time that the stress generated by radiation defects produced during the ion implantation of hydrogen into silicon promotes the formation of hydrogen-related defects. The stress and resultant strain facilitate the nucleation and growth of hydrogen platelets on planes normal to the ion implantation direction.

Using a combination of H ion implantation and optical spectroscopy we experimentally showed the relationship between H and donor-bound excitons in ZnO. H implantation into ZnO changed the relative intensities of the luminescence from the shallow donor bound excitons, enhancing the 3.361 eV peak, while IR analysis showed the formation of O-H bonds and the presence of interstitial H in a bond-centered site. These data show that implanted H becomes chemically bonded to Zn vacancies in the ZnO lattice and acts as a shallow donor.

Program impact: Provides fundamental insight at the microscopic and atomistic levels on how ion-solid interactions enhance the properties and functionality of materials. This work has enabled the synthesis of novel and improved materials.

Interactions: J. Mayer (ASU), S. S. Lau (UCSD), P. Chu (City U, Hong Kong), D. Lucca (Oklahoma State); *Internal LANL:* R. Hoagland, A. Misra, M. Baskes, J.G. Swadener, Q. Jia, J.K. Lee, L.G. Jacobsohn, D.W. Cooke

Recognitions, Honors and Awards: M. Nastasi: Fellow of LANL (2000), Recipient LANL Fellows Prize (1995), R&D 100 Award (1997), Exec. Officer of The Bohmische Physical Society (1997-), Edit. Board of Nuclear Instruments and Methods in Physics Research, Section B: (1997-), Principal Ed. J. of Materials Research (1997-2000), Chair MRS Bull Pubs Subcommittee (1994-1999), Member of International Meeting Committees: Ion Beam Modification of Materials (1995-) , Ion Implantation Technology (2002-), Chair of the Editorial Board of MRS Bull (1996-1999), *Adjunct Professor*, Univ. of Colorado, Arizona State Univ., Univ. of Maryland. Contributions over the life of the program (1996 – present): ~ 220 papers; 3 books and 5 edited volumes; 5 patents awarded; 4 PhDs awarded;

Personnel Commitments for FY2005 to Nearest +/- 10%: M. Nastasi (PI) 50%, M. Hollander (tech) 50%, L. Shao (postdoc) 50%,

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 541k

FY04 BA \$ 541k

FY05 BA \$ 333k

Laboratory Name: LANL
B&R Code: KC0203010

FWP and possible subtask under FWP:

Fundamentals of Hydroxide Conducting Systems for Fuel Cells and Electrolyzers

FWP Number:

SCPE974

Program Scope:

The vision of a Hydrogen Economy is compelling for reasons of improved energy security and environmental impact; however, before such a vision can be met there are major technological barriers that must be overcome. Namely, commercially viable technologies for the production (electrolyzers) and the use (fuel cells) of hydrogen must be developed. Hydroxide conducting systems can be used for either production or use of hydrogen and offer specific advantages compared to competing technology. To date investigation of hydroxide conductors for fuel cells or electrolyzers has been limited due to perceived limitations of durability, conductivity and/or carbonate formation. These areas have received limited study and to date little is known about the mechanisms or rates of the different decay processes. Our program combines experts in the area of fuel cells, synthetic chemistry and modeling to elucidate these decay mechanisms and design new chemical structures to overcome them. Our focus is on fully characterizing cationic materials (including currently employed potassium (which cannot be covalently bonded to a polymer) and tetraalkyl ammonium (which is known to degrade in contact with hydroxide)) and developing advanced cations capable of being covalently tethered and having sufficient stability and conductivity for use in electrolysis or fuel cell applications. The overall goal of the project is to provide an understanding of the fundamental processes that are important for the conductivity and durability of these materials. Physical processes of particular interest include: ion structure, ion charge distribution, interaction with water and anions, and the role of hydroxide versus carbonate.

Major Program Achievements (over duration of support):

While this program is relatively new, several results of merit have been obtained. Decay mechanisms of cations (including ammonium, sulfonium and phosphazanium) due to base attack have been identified under conditions relevant to fuel cells or electrolysis (1M hydroxide, 80C) and accelerated conditions (in contact with $\text{KN}(\text{SiMe}_3)_2$ in DMSO, or in contact with base at low water content). From these results, we have identified cationic structures that could reduce hydroxide attack due to steric hindrance and are in the process of synthesizing target structures. Our modeling work has shown significant differences of charge centers in different cations, and has been used to support and better interpret the degradation mechanisms witnessed in our chemical studies.

Program impact:

Current electrolyzers use aqueous potassium hydroxide and have many shortcomings due to the difficulties of handling a corrosive liquid. Advanced membrane designs would offer significant improvements in capital costs, engineering considerations and energy efficiency. The use of hydrogen in a Hydrogen Economy has largely been assumed to be through fuel cells as local power suppliers, converting the chemical energy of hydrogen into useable power. Traditional approaches involve using highly acidic polymer electrolytes as proton conductors. The use of hydroxide conductors as an alternative can both improve efficiency and improve materials compatibility issues, enabling the use of non-precious electrocatalysts. These findings would significantly increase the chances of an economically viable Hydrogen Economy and improve both US energy security and environmental concerns such as CO₂ emissions. Additionally these materials would likely be useful for other chemical processes such as catalysis, ion exchange and separation.

Interactions:

None external – program involves researchers from Materials Science, Chemistry and Theoretical Divisions.

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

None yet

Personnel Commitments for FY2005 to Nearest +/- 10%:

Bryan Pivovar: 20%; Dilip Asthigiri: 50%; Lawrence Pratt: 20%; David Thorn: 20%; Thomas Cameron/Post-Doc TBD: 100%

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 0

FY04 BA \$ 0

FY05 BA \$ 400k

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0203010

FWP and possible subtask under FWP:

Cooperative Phenomena in Molecular Nanocomposites.

FWP Number:

SCPE896

Program Scope:

The goal of this program is to create and study hybrid organic/inorganic and biological/inorganic materials in which cooperative self-assembly processes influence the formation, structure and functions of the materials. Expertise in sample preparation and characterization is applied in an integrated manner to develop new insight into the balance between forces that govern the organization and responses of the materials studied. The program is performed collaboratively with researchers at Sandia National Laboratories.

Major Program Achievements (over duration of support):

Spectroscopic characterization of surfactant/silica mesophase nanocomposite films under controlled uv-light exposure. Masked photolytic removal of organic phase in self-assembled systems and organic/inorganic nanocomposite materials to generate patterned functionalities in thin films. Developed methods for incorporation of proteins into nanoporous inorganic materials to create spatially patterned bio-composite thin films. Used neutron reflectivity to study the structure of phospholipid membrane assemblies formed on nanoporous and nanocomposite thin films. Developed methods of functionalizing inorganic nanocomposite and nanoporous materials with active chemical and macromolecular species to provide thin films with controllable luminescence in the presence of gas phase molecules. Developed bio-compatible sol-gel procedures for encapsulation of optically active proteins sensitive to sugar molecules.

Program impact:

This project has resulted in work published in high impact journals, such as *Nano letters*, *J. Phys. Chem. B*, and *Langmuir*. Developed a detailed understanding of organic template removal processes from organic/inorganic nanocomposite materials using photolytic methods. Development of spatially patterned biological/inorganic composite thin-film materials demonstrating increased stability and robustness of the biological components. Confinement and configuration of molecular and macromolecular assemblies within rigid silica frameworks has produced materials with novel optical properties that can be controlled by exposure to chemical species, with potential uses in sensor technologies. Immobilization of optically active proteins in biologically tolerant sol-gels has potential application in glucose monitoring. Development and characterization of substrate-supported phospholipid membrane assemblies has uses in biotechnology applications.

Interactions:

Atul N. Parikh (U.C. Davis); Alan Burns, Darryl Sasaki, Bruce Bunker (Sandia National Laboratories, Albuquerque); C. Jeffrey Brinker (Sandia National Laboratories, Albuquerque and the University of New Mexico); Jarek Majewski (LANSCE); Frank V. Bright (University of Buffalo); Gary A. Baker (Oak Ridge National Laboratory); Jon D. Dattelbaum (University of Richmond)

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

Symposium Organizer for Spatially Resolved Characterization of Local Phenomena in Materials and Nanostructures, Materials Research Society Fall 2002 meeting (A. Shreve, 2002); Symposium Organizer for Developing Nano-Bio Interfaces, Materials Research Society Spring 2005 meeting (A. Shreve, 2005);

Personnel Commitments for FY2005 to Nearest +/- 10%:

A. Shreve (10%); A. Dattelbaum (10%)

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 655k

FY04 BA \$ 283k

FY05 BA \$ 85k

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0203010

FWP and possible subtask under FWP:

Molecularly Engineered Biomimetic Nanoassemblies

FWP Number:

SCPE951

Program Scope:

The program aims to develop an increased understanding of fundamental interactions that govern complex molecular and biomolecular assemblies, and to explore how structure, dynamics and function are linked in such assemblies. Areas of emphasis include the development of bio-inspired assemblies with energy transduction functions, control of optical or electronic responses in complex assemblies, and exploration of the role of dynamics in controlling assembly structures and functions. The approach used includes a combination of materials synthesis and fabrication, static and time-resolved spectroscopic characterization, optical and scanning probe microscopies, neutron reflectivity, and theory and modeling of electronic responses and assembly structures.

Major Program Achievements (over duration of support):

Studied the structure and properties of several different types of bio-inspired thin-film assemblies with electronic, optical or biological responses. Examples include Langmuir-Blodgett assemblies of conjugated amphiphilic molecules that mediate photo-induced charge transfer between layers of conjugated polymers and LB-deposited amphiphilic fullerene derivatives, and photoconductive thin films based on assembly of amphiphilic quaterthiophene derivatives. Studied the electronic and optical properties of amphiphilic organic and organometallic oligomers of conjugated phenylene acetylene compounds. Used phospholipid assembly methods to create patterned hybrid and supported bilayers, and characterized lateral mobility and the effects of spatial confinement within such structures. Demonstrated that phospholipid bilayers and monolayers can be formed on electronically active surfaces such as fullerenes and modified fullerenes, and that the resulting assemblies can be characterized using optical spectroscopy, microscopy and neutron reflectivity. Demonstrated the use of protein recognition strategies to assist in the formation of controlled multi-layer assemblies of phospholipids, and carried out characterization studies using neutron reflectivity and atomic force microscopy. Controlled the luminescence properties of water-soluble conjugated polymers through the use of polyelectrolyte interactions in complex solutions, and characterized the polymer's luminescent behavior using static and time-resolved spectroscopy and density-based material separation techniques. Explored how co-assemblies of polyelectrolytes in solution can influence the photo-induced charge-transfer behavior of charged donors and neutral or charged acceptors. Synthesized a lipidic affinity peptide for fullerenes and selected peptides from combinatorial peptide library that bind to a specific conjugated polymer, enabling bio-inspired routes for complex assembly formation.

Program impact:

The program has generated improved understanding of how to control complex self assembly to create functional nanoscale materials. Applications of such materials are found in energy production and storage, catalysis, and sensor technologies.

Interactions:

Atul N. Parikh (U.C. Davis); James A. Brozik, Deborah Evans (University of New Mexico); Darryl Sasaki, John Shelnutt (Sandia National Laboratories); Jarek Majewski (LANSCE)

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

Organizer for Developing Nano-Bio Interfaces Symposium, Materials Research Society Spring 2005 meeting (A. Shreve, 2005); Member editorial board for Nanotechnology (J. Shelnutt); Invited presentations by key investigators (Shreve, Parikh, Wang) at national and international meetings.

Personnel Commitments for FY2005 to Nearest +/- 10%:

A. Shreve (30%); J. Martinez (30%); A. Dattelbaum (10%); M. Brown (postdoc, 100%); Y. Gao (postdoc, 100%); E. Batista (postdoc, 40%); H.-L. Wang (10%); R.L. Martin (10%); T. Lookman (20%); S. Iyer (20%); R. Hicks (student, 30%).

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 645k

FY04 BA \$ 645k

FY05 BA \$ 750k

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0201020

FWP and/or possible subtask under FWP:

Radiation Damage Effects In Ceramics And Non-Metals

FWP Number: SCPE429

Program Scope: The goal of this program is to understand the radiation damage response of ceramics exposed to neutrons, ions or other energetic particles. Our studies of the damage response of ceramics address two objectives: (1) to predict microstructural evolution in ceramics exposed to radiation; and (2) to identify the physical aspects of ceramics that are effective in promoting radiation resistance. Our ultimate goal is to design new radiation resistant ceramics. We conduct particle irradiation tests on ceramics to evaluate their irradiation damage response. We also perform computer simulations of damage evolution in ceramics to assist in our understanding of radiation damage phenomena in these materials. Our research is focused on highly radiation-resistant ceramics.

Major Program Achievements (over duration of support):

Using atomistic computer simulation techniques along with ion beam irradiation experiments on selected oxides, we have demonstrated that certain compounds with crystal structures similar to that of the mineral fluorite are highly resistant to displacive radiation damage. This radiation damage tolerance is very dependent on compound chemistry. For instance, our findings suggest that a large number of isochemical $A_2B_2O_7$ compounds with a structure called pyrochlore (very similar to the fluorite structure, but more highly ordered) should not exhibit resistance to radiation damage, while other $A_2B_2O_7$ compounds should be highly radiation tolerant. The radiation tolerant $A_2B_2O_7$ compounds with the fluorite structure should be suitable hosts for actinide species (Th, U, Pu, etc.). This research has been published in Science: K. E. Sickafus, L. Minervini, R. W. Grimes, J. A. Valdez, M. Ishimaru, F. Li, K. J. McClellan and T. Hartmann, "Radiation tolerance of complex oxides," Science **289** (2000) 748-751. Most recently, we have combined density functional theory, molecular dynamics, and temperature accelerated dynamics, to demonstrate how irradiation-induced defects evolve over long timescales (the oxides MgO and $MgAl_2O_4$ were chosen as a model oxides for calculation purposes).

Program Impact:

Early work on this program laid the foundation for our current understanding of the behavior of model ceramic oxides (such as periclase (MgO) and corundum (Al_2O_3)) in a radiation damage environment. Recent developments on this program have led to an exciting new predictive capability to predict radiation damage evolution in model oxides such as MgO to time scales approaching experimental (order of seconds). Other efforts: Kurt Sickafus organized an international symposium on "Spinel Compounds: Structure Property Relations" at the Annual Meeting of the American Ceramic Society, Cincinnati, OH (1998). The proceedings was published as a special topical issue of the Journal of the American Ceramic Society (Vol. 82(12) 1999). Kurt Sickafus served as Director of a NATO-ASI international school entitled "Radiation Effects in Solids," held in Erice, Sicily in July, 2004. Kurt served as chair the 13th International Conference on Radiation Effects in Insulators, held in Santa Fe, NM, Aug. 28 - Sept. 02, 2005. Kurt Sickafus and Blas Uberuaga are serving as Guest Editors (NIM-B) for the proceedings of this conference.

Interactions:

R. W. Grimes (Imperial College); R. Smith (Loughborough University); M. Ishimaru (Osaka University); K. Yasuda (Kyushu University); H. Matzke (Institute for Transuranic Elements, Karlsruhe, GERMANY); V. T. Gritsyna (Kharkiv State University, Kharkiv, UKRAINE). *Internal LANL Interactions:* Art Voter, Steve Valone, Marius Stan, Chris Wetteland, and Mike Baskes.

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

Fellow of the American Ceramic Society to Frank Clinard (1986)
Foreign Distinguished Visiting Scientist at JAERI, Tokai, JAPAN to Frank Clinard (1987)
Los Alamos National Laboratory Fellow's Prize to Mike Nastasi (1995)
Fellow of the American Ceramic Society to Kurt Sickafus (1998)
Fellow of Los Alamos National Laboratory to Mike Nastasi (2000)
Los Alamos National Laboratory Fellow's Prize to Kurt Sickafus (2001)
2002 OBES Chunky Bullet competition co-winner (2002)

Personnel Commitments for FY2005 to Nearest +/-10%:

K. Sickafus (100%), J. Valdez (100%), B. Uberuaga (50%), M. Tang (100%), A. Cleave (100%), (50%, D. Bacorisen).

Authorized Budget (BA) for FY03, FY04, FY2005:

FY03 BA \$ 843 k

FY04 BA \$ 843k

FY05 BA \$ 809k

Laboratory Name: LANL
B&R Code: KC0201030

FWP and possible subtask under FWP:

Electronic Processes in Solid State Organic Electronic Materials

FWP Number: SCPE973

Program Scope:

The field of organic electronic materials is in a state of rapid expansion. There are strong technology drivers for this field but to date much of the work in this research area has employed Edisonian approaches. There is a very large material and device structure phase space in this field and we must move to a predictive science based approach. Organic electronic materials are condensed phases of p-conjugated molecules and intermolecular interactions are critical in determining the condensed phase properties. The electronic properties of the dense films are not the same as those of the isolated molecules or single polymer chains. A major theme of this proposed research is aimed at determining which features of the electronic structure of the isolated molecules are maintained and which are modified in the condensed phase and to use our understanding of molecular electronic structure to describe the behavior of the condensed phases. The overall goal of the project is to provide an understanding of the fundamental physical processes that are important in determining the properties of organic electronic materials. Physical processes of particular interest include: electrical injection, charge transport, electron spin injection, transport and dynamics and exciton dynamics and transport.

Major Program Achievements (over duration of support):

π -conjugated organic semiconductors are promising for non-equilibrium electron spin based phenomena because they have extremely small spin-orbit interactions and small hyper-fine interactions so that both electron and hole spin lifetimes are long. Organometallic compounds with strong spin-orbit interactions can be included at specific spatial locations in a device structure to permit manipulation of spin-orbit coupling without adverse effects on spin lifetimes elsewhere in the structure. We measured magnetic field effects on the photoluminescence of organometallic molecules in organic matrices. We identified promising organometallic compounds for studies of manipulation and control of spins in organic materials and devices. We found that the photoluminescence (PL) of the organometallic molecule PtOEP in a blue polymer matrix (PFO) showed strong magnetic field dependence. The highest energy peak of the PtOEP PL spectrum diminished as the temperature dropped to 2K. Application of a magnetic field brought back this high energy peak. We studied the circular polarization of PL from PtOEP at fields up to 40 T for unpolarized excitation. We found that the emission was 30% polarized. We are pursuing using this molecule for optical injection of spin into organic semiconducting materials.

Program impact:

Organic electronic materials are of great intrinsic scientific interest. In these materials there is strong coupling of electron charge, spin, and lattice degrees of freedom, and the rich spectrum of tunable ground and excited states that results make them an ideal vehicle for the study of flexible, strongly correlated systems. This system richness is enhanced by the wide variety of material and structural properties available through synthetic chemistry and the ability to fabricate nanoscale structures through molecular assembly techniques. The magneto-optic measurements are demonstrating the importance of spin-lattice coupling in organic semiconductors.

Interactions:

Established collaboration with National High Magnetic Field Laboratory for magneto-optical measurements.

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

none

Personnel Commitments for FY2005 to Nearest +/- 10%:

Ian Campbell: 20%

Brian Crone: 20%

Rich Martin: 20%

Darryl Smith: 30%

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 0

FY04 BA \$ 0

FY05 BA \$ 450k

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0202020

FWP and possible subtask under FWP:

Science of Electronic and Optical Interactions between Coupled Nanostructures

FWP Number: SCPE395

Program Scope: The goal of this project is the understanding of the behavior of interacting nanophotonic and nanoelectronic structures. One focus area is the interactions that produce changes in the optical and electronic properties of the nanostructures. We are investigating the effect of inter-dot interactions on the optical and electronic properties of self-assembled and colloidal quantum dot arrays. The second activity will be an exploration of novel composite materials consisting of semiconductor colloidal quantum dots embedded in Inorganic and organic semiconductors. A second focus area is the study of nanoelectronic structures in photonic bandgap materials and photonic crystal fibers. The interplay between the restricted electronic density of states and the restricted photonic density of states can produce large optical nonlinearities, can inhibit radiative decay, and can perhaps lead to high speed optical switching.

Major Program Achievements (over duration of support): We explored the effect of semiconductor nanocrystal (NC) “geometry” on the rates of the multi-particle decay. In particular, we study the influence of the zero- to one-dimensional (1D) transformation on multi-particle Auger recombination using a series of elongated semiconductor NCs (quantum rods). We observe the transition from the three- to two-particle recombination process as the nanocrystal aspect ratio is increased. This transition implies that in the 1D confinement limit, Auger decay is dominated by Coulomb interactions between 1D excitons that recombine in a bimolecular fashion. One consequence of this effect is strongly reduced decay rates of higher multi-particle states that lead to increased optical gain lifetime and efficient light amplification due to transitions involving excited electronic states. These unique rod properties suggest that shape control may be key to developing practical lasing applications of NCs.

We explored the use of NC core-shell heterostructures for suppressing Auger recombination. Specifically, we use inverted ZnSe(core)/CdSe(shell) nanoparticles to study the effect of type I vs. type II carrier localization on multiparticle Auger decay and optical gain performance of NCs. We observe that both Auger recombination rates and the amplified spontaneous emission thresholds are reduced in the case of type II localization. Furthermore, we use these hetero-NCs to demonstrate efficient amplified spontaneous emission that is tunable across a “difficult” range of green and blue colors.

As a method for indirect charge injection into nanocrystals, we explored the use of energy transfer from a shallow quantum well. For these studies, we fabricated *GaInN* quantum wells with a thin top barrier. We used Langmuir-Blodgett techniques to assemble a monolayer of nanocrystals on top of the quantum well. We investigated exciton dynamics in the quantum well with and without the nanocrystal monolayer. This comparison indicates an extremely efficient exciton transfer (~50%) from the quantum well to the NQDs. These experiments represent the first direct observation of Coulomb coupling between epitaxial and colloidal nanostructures.

We have studied a wide range of nonlinear-optical effects in photonic crystal fibers (PCF). These waveguides have nanoscale structure of glass and/or other materials in the transverse plane (perpendicular to the optical field propagation direction) and are invariant along the length (propagation direction). Said nanostructure strongly alters linear (dispersion) as well as nonlinear waveguiding properties of the fiber. Among the effects studied are supercontinuum generation in silica and SF₆ glass PCF; phase-matched third harmonic generation; very high-order UV mode emission; soliton dynamics, stabilization and Cherenkov continuum generation; resonant scattering of continuous waves on solitons. A unique set of tools and capabilities were developed (XFROG, imaging, numerical models) and will be used in future work.

Program impact: This project addresses important scientific issues at the forefront of nanoscience. It complements ongoing R&D activities ongoing at the Center for Integrated Nanotechnologies (CINT). The portfolio of activities contained within this program will unify nanoelectronics and nanophotonics research at Sandia and Los Alamos, further establishing a firm foundation of nanoscience relevant to CINT.

Interactions: Department of Physics, University of Bath, UK; Department of Physics, University of Florida, Gainesville; Department of Chemistry, MIT; Department of Physics, Georgia State University

Recognitions, Honors and Awards: V.I. Klimov—Fellow of the American Physical Society, (2003), Fellow of the Optical Society of America, (2003), Los Alamos Laboratory Fellow (2004), 14 invited talks

Personnel Commitments for FY2005 to Nearest +/- 10%:

Staff: A. Efimov, 30%, V.I. Klimov, 5%; postdocs H. Htoon, 50%

F FY03 BA \$ 210k

FY04 BA \$ 204k

FY05 BA \$ 190k

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0202010

FWP and possible subtask under FWP:

Complex Electronic Materials

FWP Number: SCPE355

Program Scope: Research focuses on developing a fundamental understanding of the physics of complex and collective states in electronic materials by discovering new materials that reveal essential new physics. A necessarily broad range of experimental techniques, often at extremes of low temperatures, high fields and high pressures, is used to probe static and dynamic spin, charge and lattice degrees-of-freedom and their interactions on multiple length and time scales. Particular attention is given to highly correlated f-electron materials and layered cuprates, often in single crystal form, as exemplary complex electronic materials.

Major Program Achievements (over duration of support): Established the study of heavy-fermion materials as a new field of condensed matter physics through discoveries of numerous examples of unconventional superconducting, magnetic and semiconducting states in new correlated f-electron systems; discovered unconventional superconductivity in PuCoGa_5 and in a new family of $\text{Ce}_n\text{MIn}_{3n+2}$ ($\text{M}=\text{Co}, \text{Rh}, \text{Ir}$; $n=1,2$) materials; discovered high T_c superconductivity in rare-earth cuprates, providing the first indication for the importance of CuO_2 planes; pioneered the now widely accepted importance of intrinsic inhomogeneity in the spin, charge and lattice of cuprates, using neutron and NMR spectroscopies; in CeCoIn_5 , discovered evidence for Fulde-Ferrel-Larkin-Ovchinnikov state, first predicted nearly 40 years ago, and for an unusual form of quantum criticality; discovered superconductivity in hole-doped diamond; discovered a field-induced line of magnetic quantum-critical points within the pressure-induced superconducting state of CeRhIn_5 .

Program Impact: Program is recognized internationally for its leadership in creating new science through the discovery and study of new correlated electron materials.

Interactions: Z. Fisk (U. C. Davis), D. E. MacLaughlin (U.C. Riverside), B. Buchner (University Cologne), C. Retorri (UNICAMP), J. M. Lawrence (U.C. Irvine), Y. Kitaoka (Osaka University), M. Nicklas (Max-Planck Institute for the Chemical Physics of Solids), P. Oppeneer (Uppsala University), G. Lander (ITU, Karlsruhe), V. Sidorov (IHPP, Troitsk), L. Greene (University of Illinois), Q. Si (Rice University) among many others.

Recognitions, Honors and Awards (at least partially attributable to support under this FWP or subtask): Z. Fisk-National Academy of Science, American Academy of Arts and Sciences, E. O. Lawrence Prize, DOE Award for Sustained Outstanding Research in Solid State Physics, APS New Materials Prize, APS Fellow, APS Div. Condens. Matt. Phys. Executive Committee, APS Buckley Prize Committee, LANL Fellow, editorial boards of *Physica B* and *Phys. Rev. Lett.*; P. C. Hammel-APS Fellow, LANL Fellow, LANL Fellows' Prize, APS Executive Committee of Instrumentation and Measurement Science; R. H. Heffner-APS Fellow; R. Movshovich-LANL Fellows' Prize ; R. Pynn-APS Fellow, AAAS Fellow; J. L. Sarrao-LANL Fellow's Prize; J. L. Smith- E. O. Lawrence Prize, DOE Award for Sustained Outstanding Research in Solid State Physics, APS New Materials Prize, APS Fellow, LANL Fellow, editorial boards of *J. Alloys and Compds.* and *Phil. Mag.*; J. D. Thompson- APS Fellow, AAAS Fellow, APS Div. Condens. Matt. Phys. Executive Committee, DOE Award for Sustained Outstanding Research in Solid State Physics, ISI Highly Cited Physicist, LANL Fellow, LANL Fellows' Prize, Japanese Society for the Promotion of Science Fellow; collectively served on numerous International Conference Advisory Committees and review panels, and within past year presented over 20 invited talks at international conferences/workshops

Personnel Commitments for FY2005 to Nearest +/-10%: J. D. Thompson (15%), W. Bao (35%), N. Curro (20%), R. Movshovich (20%), J. L. Sarrao (15%), three postdocs (75%)

Authorized Budget (BA) for FY03, FY04, FY05

FY03 BA \$1,205,000

FY04 BA \$850,000

FY05 BA \$880,000

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0201020

FWP and possible subtask under FWP:

Ensemble-Controlled Deformation Behavior in Materials

FWP Number: SCPE401

Program Scope: Our primary goal is to use an integrated experimental and theoretical approach to characterize the complex role played by ensembles of dislocations, twins, and grain boundaries during plastic deformation. The scope of the proposed work is to connect the various length scales and physical mechanisms governing deformation (micro, meso and macro scale), in order to explain the overall mechanical response of metallic aggregates. Starting in FY03 the focus of this program has been hexagonal materials, where the coupling between the microscopic mechanisms and the macroscopically observed response is very strong, and not completely understood. The paradigm of this program is to reveal such coupling by studying complex testing histories involving changes in strain path and/or temperature and/or strain rate

Major Program Achievements (over duration of support): This program uses as a starting point the developments achieved under the previous Mechanical Properties Program. Specifically: use polycrystal constitutive laws to account for plastic response and anisotropy in terms of crystallographic texture and crystallographic features, for a wide range of strain, temperature and strain-rate regimes.

Recently we have incorporated experimental information about the grain microstructure and its evolution into our polycrystal models. Specifically, we have developed experiment-based models for dislocation walls, intragranular banding, misorientation, and twin domains. We have applied this integrated approach to cubic materials and, in the last three years, also to hexagonal materials, with emphasis in Zr and Mg. We are making intensive use of in-house neutron diffraction facilities for characterizing microstructure, and we have developed new automated or combined techniques for performing TEM and OIM analysis. We have developed new Multiple-State Embedded Atom Potentials for a basic understanding of dislocation-twin interactions. And we have developed more physically-based polycrystal constitutive models for describing the mechanical response of HCP aggregates.

Program impact: The effort on texture, anisotropy, constitutive description and modeling of polycrystal plasticity came at a time when the Materials Science community started becoming aware of these issues. It changed the way in which constitutive modeling was approached by the scientific community, and Los Alamos is regarded as leading the field in such issues. Today the field has matured, and the scientific community is looking at the specific role played by defects such as dislocations, grain boundaries and twins on the overall mechanical response of polycrystals. Such quest, which can only be answered through a basic and integrated study of such features, is the general scope of this program. We predict that our specific focus on hexagonal aggregates, such as Zr and Be alloys, will have a direct impact on basic material science and in Los Alamos research programs. In addition, the study of Ti and Mg alloys will impact energy related technologies. As a spin-off, we foresee an impact in the understanding of systems where phase transformations are important, such as piezo-electric and shape-memory alloys.

Interactions: A. J. Beaudoin (U of IL-Urbana), I. Robertson (U. of IL-Urbana), D. Embury (McMaster U.), S. I. Wright (TexSEM Labs), A. H. Heuer (CWRU), J.P. Hirth (LANL consultant), S.R. Agnew (U. of Virginia), S. Kalidindi (Drexel U.), W. Horstemeyer (MS State U.), P. van Hove (Law. Berk. NL), C.P Wong (GA Inst. Tech.), R. Holt (Queen's U.), B-J. Lee (Pohang U. Japan), R. Asta (Northwestern U.)

Recognitions, Honors and Awards (at least partly attributable to support under this FWP): U. Fred Kocks, Fellow, (NAE, TMS, LANL, ASM, APS, AIME); T.E. Mitchell, Fellow (LANL, TMS, ASM, APS, ACS), Honorary D. Sc., U. of Cambridge; Symp. and special issue of Phil. Mag. A in his honor; M.I. Baskes, Fellow (TMS, LANL, IP), DOE BES Award for Sustained Outstanding Research, Journal Editor, *Modeling and Sim. in Materials Sci. and Eng.*; About 30 Invited Presentations (altogether in the last three years) related to this project. About 20 papers published in International peer reviewed journals in the last three years related to this project.

Personnel Commitments for FY2005 to Nearest +/- 10%: C. Tomé (PI, 50%), M. Baskes (20%), I. Beyerlein (30%), G Kaschner (50%), A Misra (20%), S. Srivilliputhur (50%), R. McCabe (30%), E. Cerreta (20%), B. Clausen (30%), G. Proust (100%, post-doc), S. Li (30%, post-doc), A. Jain (100%, PhD student).

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$1,043,000

FY04 BA \$1,043,000

FY05 BA \$1,001,000

Laboratory Name: Los Alamos National Laboratory
B&R Code: KC0202030

FWP and/or subtask Title under FWP:
Accelerated Molecular Dynamics Methods

FWP Number: SCPE420

Program Scope:

The goals of this program are to develop methods for extending the time scale of molecular dynamics simulations to reach times relevant for diffusive processes and experiments; to improve the methods to make them applicable to the widest possible range of processes in chemistry, physics, materials science, nanotechnology, and biology; to apply the methods to problems of interest to LANL and DOE; and to collaborate with groups around the world to apply the methods to key problems in which they are expert.

Major Program Achievements (over duration of support):

Developed the parallel-replica dynamics method, which achieves parallel speedup for infrequent-event systems while maintaining exact dynamics. Developed the temperature accelerated dynamics (TAD) method, which achieves significant boost (many orders of magnitude) when barriers are high relative to the temperature. Used MD+TAD to simulate vapor-deposited metallic crystal and film growth at experimental deposition rates (seconds per monolayer), observing importance of highly concerted activated processes for surface smoothing even at low temperatures. Studied interstitial diffusion of H₂ in fcc fullerene lattice, discovering unexpected multiple-occupancy mechanism. Performed first study of low-energy radiation damage and annealing on experimental time scales (seconds) with full atomistic detail (MgO).

Program impact:

The accelerated molecular dynamics concept, that the best way to evolve a system from state to state is to let the trajectory find its own way out of each state, is impacting the way people view infrequent-event systems and activated processes. Accelerated molecular dynamics simulations are elucidating key mechanisms in processes such as surface diffusion, vapor-deposited crystal growth, bulk diffusion, radiation damage annealing, and carbon nanotube dynamics.

Interactions:

LANL internal: ASCI Enhanced Surveillance program, Advanced Fuels (AFCI) program, BES ceramics radiation damage simulation program. *External:* Jacques Amar (University of Toledo), Luciano Colombo (University of Cagliari, Italy), Jimme D. Doll (Brown University), Riccardo Ferrando (University of Genova), John Harding (University College London, UK), Robin Grimes (Imperial College, London), John Hamilton (Sandia National Laboratory, California), Graeme Henkelman (University of Texas), Hannes Jónsson (University of Iceland), Janna Maranas (Pennsylvania State University), Yuri Mishin (George Mason University), Francesco Montalenti (University of Milano/Bicocca, Italy), David Sholl (Carnegie Mellon University), Roger Smith (University of Loughborough, UK), James Sprague (Naval Research Laboratory), David Srolovitz (Princeton), Steve Stuart (Clemson University), Greg Voth (University of Utah), John Wilkins (Ohio State), Wolfgang Windl (Ohio State University).

Recognitions, Honors and Awards (at least partly attributable to support under this FWP or subtask):

A.F. Voter (PI):

Los Alamos Laboratory Fellow (2003)

Nominated to editorial boards of Journal of Chemical Physics and Theoretical Chemistry Accounts

Numerous invited talks at national and international conferences (e.g., ACS, APS, MRS, TMS, AIChE, E-MRS)

Numerous invitations to write articles and book chapters

Personnel Commitments for FY2005 to Nearest +/- 10%:

A.F. Voter 90%

Postdocs and collaborators funded from other sources

Authorized Budget (BA) for FY03, FY04, FY05:

FY03 BA \$ 360k

FY04 BA \$ 360k

FY05 BA \$ 340k